

PATENT ABSTRACTS OF JAPAN

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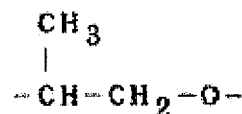
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(54) CURABLE COMPOSITION

(57)Abstract:

PROBLEM TO BE SOLVED: To provide curable compositions having excellent weatherability and heat resistance.

SOLUTION: Curable compositions comprise an oxypropylene polymer whose polymer main chain has repeating units represented by the formula and which has at least one silicon atom-containing group having a silicon atom bonded to a hydroxyl group or a hydrolyzable group, an Mw/Mn of not greater than 1.6, and a number average molecular weight of not smaller than 6,000 and a hindered phenol based or hindered amine based antioxidant.



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CLAIMS

[Claim(s)]

[Claim 1](A) A polymerization main chain. [Formula 1]

$$\text{CH}_3$$
$$|$$
$$\text{式 } -\text{CH}-\text{CH}_2-\text{O}-$$

It is an oxypropylene polymer which has at least one silicon atom content group containing the silicon atom which came out, and contained the repeating unit shown and the hydroxyl group or the hydrolytic basis combined. The hardenability constituent in which Mw/Mn contains the oxypropylene polymer whose number average molecular weight is 6,000 or more, and the (B) antioxidant or less by 1.6.

[Claim 2](A) The hardenability constituent according to claim 1 whose Mw/Mn of a polymer of an ingredient is 1.5 or less.

[Claim 3](A) The hardenability constituent according to claim 1 or 2 whose number average molecular weights of a polymer of an ingredient are 6,000-30,000.

[Claim 4](A) A hardenability constituent given in any 1 paragraph of Claims 1-3 to which a silicon atom content group exists in molecular chain terminals in a polymer of an ingredient

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the oxypropylene polymer containing a reactive silicon group, and the new hardenability constituent containing an antioxidant.

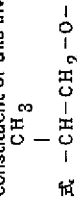
[0002]

[Description of the Prior Art] The oxypropylene polymer which has a reactive silicon group (basis which is a silicon atom content group containing the silicon atom which the hydroxyl group or the hydrolytic basis combined, and can form a siloxane bond) can turn into a liquefied polymer, is hardened at a room temperature with hygroscopic surface moisture etc., and produces a rubber-like hardened material. For this reason, this polymer is used for the elastic sealant of the building, etc. When using this polymer, in order to improve weatherability, heat resistance, etc. of a hardened material, it may be used as a constituent which added the antioxidant.

[0003] As a result of examining the hardenability constituent containing the oxypropylene polymer which has the reactive silicon group which was excellent in weatherability, when the narrow thing of molecular weight distribution was used as an oxypropylene polymer, this invention persons found out that weatherability and heat resistance were improved further, and resulted in this invention.

[0004]

[Means for Solving the Problem and its Function] (A) polymerization main chain a hardenability constituent of this invention, [Formula 2]



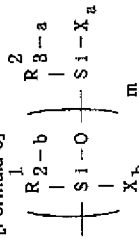
It is an oxypropylene polymer which has at least one silicon atom content group (reactive silicon group) containing a silicon atom which came out, and contained a repeating unit shown and a hydroxyl group or a hydrolytic basis combined, Mw/Mn (weight average molecular weight/number average molecular weight) contains an oxypropylene polymer whose number average molecular weight (Mn) is 6,000 or more or less in 1.6, and the (B) antioxidant.

[0005]

[Embodiment of the Invention] Although the reactive silicon group in particular as used in the field of this invention is not limited, if a typical thing is shown, a following general formula and the basis expressed with ** 3 will be mentioned, for example.

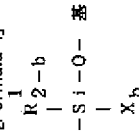
[0006]

[Formula 3]



R¹ and R² among [type, All The alkyl group of the carbon numbers 1-20, the aryl group of the carbon numbers 6-20, When the Tori ORGANO siloxy group shown by the aralkyl group of the carbon

numbers 7-20 or (R¹)₃SiO- is shown and R¹ or two or more R² exist, they may be the same and may differ. R¹ is a univalent hydrocarbon group of the carbon numbers 1-20 here, and three R¹ may be the same and may differ. X shows a hydroxyl group or a hydrolytic basis, and when two or more X exists, they may be the same and may differ. As for a, 0, 1, 2, or 3b shows 0, 1, or 2, respectively, m pieces [Formula 4]



b which can be boiled and set may differ. m shows an integer of 0-19. However, a+sigma b>=1 shall be satisfied. J.

[0007] A hydrolytic basis in particular shown by the above X is not limited, but should just be a conventionally publicly known hydrolytic basis. Specifically, a hydrogen atom, a halogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an acid-amide group, an aminoxy group, a sulfinyl group, an alkenyloxy group, etc. are mentioned, for example. Among these, although a hydrogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an aminoxy group, a sulfinyl group, and an alkenyloxy group are preferred, especially alkoxy groups, such as a viewpoint of hydrolysis nature being quiet and being easy to deal with it to a methoxy group, are preferred.

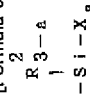
[0008] 1-3 of this hydrolytic basis and hydroxyl group can be combined with one silicon atom, and, as for (a+sigma m), it is preferred that it is 1-5. When a hydrolytic basis and a hydroxyl group exist in [two or more] a reactive silicon group, they may be the same and may differ.

[0009] Although there may be one silicon atom and there may be two or more pieces in a reactive silicon group, in the case of a reactive silicon group with which a silicon atom was connected by siloxane bond etc., there may be about 20 pieces.

[0010] A reactive silicon group expressed with a following general formula and ** 5 --- acquisition --- from an easy point, it is desirable.

[0011]

[Formula 5]



(R², X, and a are the same as the above among a formula.)

[0012] As an example of R¹ in the above-mentioned general formula and ** 3, and R², For example, cycloalkyl groups, such as alkyl groups, such as a methyl group and an ethyl group, and a cyclohexyl group, Aralkyl groups, such as aryl groups, such as a phenyl group, and benzyl, the Tori ORGANO siloxy group R¹ is indicated to be by ₃SiO- which is a methyl group, a phenyl group, etc. (R¹), etc. are mentioned. Especially as R¹, R², and R¹, a methyl group is preferred.

[0013] A reactive silicon group is good for 1.1-5 pieces to exist preferably in [at least one] one molecule of oxypropylene polymers. If the number of the reactive silicon groups contained in one molecule of polymers will be less than one piece, hardenability will become insufficient and will become difficult to reveal a good rubber elasticity action.

[0014] A reactive silicon group may exist in an end of an oxypropylene polymer chain, and may exist in an inside. Since effective network chain density of an oxypropylene polymer component contained in a hardened material formed eventually will increase if a reactive silicon group exists in an end of a chain, a rubber-like hardened material in which a low elastic modulus is shown becomes is easy to be obtained by high intensity and high elongation.

[0015] An oxypropylene polymer which constitutes a polymerization main chain in a polymer of this invention, [Formula 6]



It comes out and the repeating unit shown is contained. This oxypropylene polymer may be straight chain shape, may be a letter of branching, or may be these mixtures. Although other monomeric units may be included, it is preferred that 80 % of the weight or more of monomeric units expressed to ** 6 exist preferably 50% of the weight or more in a polymer.

[0016] Although 6,000 or more things are used effectively and get as a number average molecular weight (Mn) of this oxypropylene polymer, what has a number average molecular weight of 6,000-30,000 is good preferably. In this oxypropylene polymer, the ratio (Mw/Mn) of weight average molecular weight and a number average molecular weight is 1.6 or less, and molecular weight distribution is very narrow (monodisperse nature is large). The value of Mw/Mn is 1.5 or less preferably, and is 1.4 or less still more preferably. Although it is measurable with various kinds of methods, the measurement of molecular weight distribution by a gel permeation chromatography (GPC) method is usually common. Thus, although a number average molecular weight is large, since molecular weight distribution is narrow, before hardening, viscosity is low, the constituent of this invention is easy handling, and a good rubber-like elasticity action is shown after hardening.

[0017] As for an oxypropylene polymer which has a reactive silicon group which is the (A) ingredient of this invention, it is preferred to obtain by introducing a reactive silicon group into an oxypropylene polymer which has a functional group.

[0018] Although molecular weight distribution is narrow in the amount of Polymer Division and it is very difficult to obtain an oxypropylene polymer which has a functional group by an elongation reaction method which used the usual polymerizing method (anionic polymerization method using a caustic alkali) and this polymer of oxypropylene as a raw material, it can obtain by a method indicated to JP,61-197631.A, JP,61-215623.A, JP,61-215623.A, JP,61-215623.A, JP,46-27250.B, JP,59-15336.B, etc. which are the special polymerizing methods. Since molecular weight distribution is in a tendency which spreads as compared with a polymer before introduction when a reactive silicon group is introduced, a thing narrow as much as possible of molecular weight distribution of a polymer before introduction is preferred.

[0019] What is necessary is just to perform introduction of a reactive silicon group by a publicly known method. That is, for example, the following methods are mentioned.

[0020] (1) Make hydrosilane which has a hydrolytic basis act on a resultant which made an organic compound which has an active group and an unsaturation group which show reactivity to an oxypropylene polymer which has functional groups, such as a hydroxyl group, to this functional group react to an end, and was acquired by ranking second, and hydrosilylate.

[0021] (2) Make a compound which has a functional group (henceforth Y functional group) and a reactive silicon group which show reactivity to an oxypropylene polymer which has functional groups (henceforth Y functional group), such as a hydroxyl group, an epoxy group, and an isocyanate group, to this Y functional group react to an end.

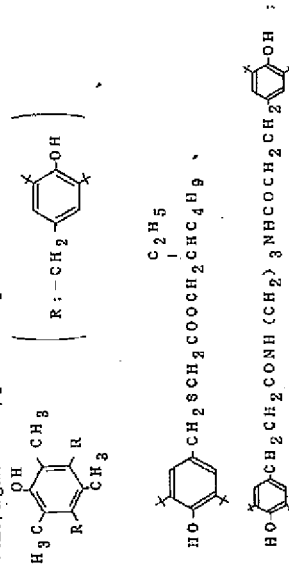
[0022] As a silicon compound which has this Y functional group, gamma-(2-aminoethyl) aminopropyl trimethoxysilane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, Amino group content Silang, such as gamma-aminopropyl triethoxysilane; gamma-mercaptopropyltrimethoxysilane, Sulfhydryl group content Silang, such as gamma-mercaptopropylmethyl dimethoxysilane; Gamma-glycidyloxypropyltrimethoxysilane, Epoxysilane, such as beta-(3, 4-epoxycyclohexyl) ethyltrimethoxysilane; Vinyltrimethoxysilane, gamma-methacryloyl oxypropyl trimethoxysilane, vinyl type unsaturation group content Silang [such as gamma-acryloyloxypropylmethyl dimethoxysilane,]; — chlorine atom content Silang [such as gamma-chloropropyltrimethoxysilane,]; — gamma-isocyanatepropyl triethoxysilane. Isocyanate content Silang, such as gamma-isocyanate propylmethyl dimethoxysilane; although hydro-Silang, such as methyl dimethoxysilane, trimethoxysilane, and methyl diethoxysilane, is illustrated concretely and they get, it is not limited to these.

[0023] Method ** which makes a compound which has a polymer, an isocyanate group, and a reactive silicon group which have a hydroxyl group react to an end a method of (1) or among (2) in the above method is preferred.

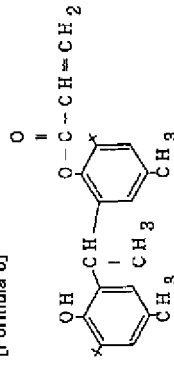
[0024] (E) It is preferred for an antioxidant which is an ingredient to use a thing of a hindered phenol

system and a hindered amine system. It is not limited by these although various things indicated to an "antioxidant handbook" of completion company issue, "degradation, stabilization" (the 235-242nd page) of the CMC Co., Ltd. issue of a polymer material, etc. are mentioned as these examples.

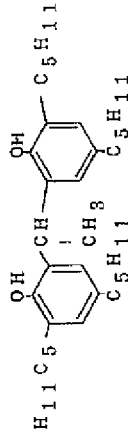
[0025] Namely, as an antioxidant of a hindered phenol system, 2,6-di-tert-butyl-4-methyl phenol, 2,6-di-tert-butyl-4-ethylphenol, Mono(or JJ or Tori) (alpha-methylbenzyl) phenol, 2,2'-methylenebis (4-methyl-6-tert-butylphenol), 2,2'-methylenebis (4-methyl-6-tert-butylphenol), 4,4'-Thiobis (3-methyl-6-tert-butylphenol), 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amyl hydroquinone, A triethylene glycol screw [3-(3-tert-butyl-5-methyl-4-hydroxyphenyl) propionate], 1,6-hexane ***** [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate], 2,4-bis-(n-octylthio)-6-(4-hydroxy-3,5-di-tert-butylanilino)-1,3,5-triazine, Pentaerythrityl tetrakis [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate], A 2,2-thio-diethylenescrew [3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate], Octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, N,N'-hexa methylenebis (3,5-di-tert-butyl-4-hydroxy-hydronalium thinner MAMDO), 3,5-di-tert-butyl-4-hydroxy-benzylphosphonate-diethyl ester, 1,3,5-trimethyl 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenyl) benzene, Bis(3,5-di-tert-butyl-ethyl 4-hydroxybenzylphosphonate)calcium, Tris-(3,5-di-tert-butyl-4-hydroxyphenyl)-isocyanurate, 2,4-screw [octylthio] methyl-O-cresol, N,N'-bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionyl] hydrazine, Tris (2,4-di-tert-butylphenyl) phosphite, 2-(5-methyl-2-hydroxyphenyl) benzotriazol, 2-[2-hydroxy-3,5-bis(alpha and alpha-dimethylbenzyl)phenyl]-2H-benzotriazol, 2-(3,5-di-tert-butyl-2-hydroxyphenyl) benzotriazol, A 2-(3-tert-butyl-5-methyl-2-hydroxyphenyl)-5-chlorobenzotriazole, A 2-(3,5-di-tert-butyl-2-hydroxyphenyl)-5-chlorobenzotriazole, A 2-(3,5-di-tert-butyl-5-(2H-benzotriazol 2-yl)-4-hydroxyphenyl) benzotriazol, A condensate with a methyl-3-[3-tert-butyl-5-(2H-benzotriazol 2-yl)-4-hydroxyphenyl] propionate polyethylene glycol (molecular weight 300 [about J], Hydroxyphenyl benzotriazole derivatives, a 2-(3,5-di-tert-butyl-4-hydroxybenzyl)-2-n-butylmalonic acid screw (1,2,2,6,6-pentamethyl 4-piperidyl), 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxy benzoate, Mark AO-30 which are the products made from ADEKAAGASU, IrganoxMD 1024 which is a product made from Mark AO-80;Ciba Geigy, Irganox 1425, Irganox 245, [Formula 7]



S. Isonox 129 which is a product made from Antioxidant HPM-12;Schenectady which is a product made from F.O.S; SamilizerGM, SumilizerGA-80 which are the Sumitomo Chemical Co., Ltd. make, [Formula 8]

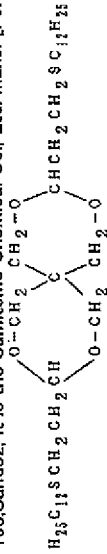


NaugardXL-1 which is a product made from Uniroyal; in addition to this, [Formula 9]



**** is mentioned.

[0026]As an antioxidant of a hindered amine system, The succinate dimethyl 1-(2-hydroxyethyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine polycondensation thing, poly — [6-(1,1,3,3-tetramethylbutyl) amino-1,3,5-triazine 2,4-diyl] and [(2,2,6,6-tetramethyl 4-piperidyl) imino] hexamethylene [(2,2,6,6-tetramethyl 4-piperidyl) amino]—6-chloro-1,3,5-triazine diamine 2,4-screw [N-butyl-N-(1,2,2,6,6-pentamethyl 4-piperidyl) amino]—6-chloro-1,3,5-triazine condensate, Bis(2,2,6,6-tetramethyl 4-piperidyl)sebacate, succinic acid-bis(2,2,6,6-tetramethyl 4-piperidyl)sebacate, which is a product made from Ciba Geigy. Mark PEP-36 which is a PIPERIDINURJester, Irgafos which is a product made from Ciba Geigy. Mark PEP-36 which is a product made from Ultrinox 626; ADEKAAGASU which is a product made from Cyanox 1790; Borg Warner which is a product made from Sandstab P-EPQ; ACC which is a product made from Sumitomo Chemical Co., Ltd. make. [Formula 10]



**** is mentioned.

[0027]Although there is no limitation in particular as amount of the above-mentioned antioxidant used, it is preferred that they are 0.01–10 copies to reactive silicon group content oxypropylene polymer 100 weight section (only henceforth a "part") which is the (A) ingredient, and it is still more preferred that they are 0.1–5 copies. An antioxidant may be used independently, and may use together and use two or more sorts. It is preferred to mix and use a thing of a hindered phenol system and a thing of a hindered amine system especially.

[0028]In stiffening a constituent of this invention, even if it uses a curing catalyst, it is not necessary to carry out. When using a curing catalyst, a publicly known thing can be used widely conventionally. As the example, titanate, dibutyltin dilaurate, such as tetrabutyl titanate and tetrapropyl titanate, Dibutyltin maleate, dibutyltin diacetate, octylic acid tin, tin carboxylate [such as naphthenic acid tin], — reactant [of dibutyltin oxide and phthalic ester]; — dibutyltin diacetyl acetonate; — aluminum tris acetylacetonate. Organoaluminum compounds, such as aluminumtrisethylacetate and diisopropoxy aluminum ethylacetate; Zirconium tetra acetylacetonate, chelate compound [such as titanium tetra acetylacetonate], — lead octylate; — a butylamine. Octylamine, lauryl amine, dibutyl amine, monoethanolamine, Diethanolamine, triethanolamine, diethylenetriamine, Triethylenetetramine, oleylamine, cyclohexylamine, Benzylamine, diethylamino propylamine, xylene diamine, triethylenediamine, guanidine, diphenylguanidine, 2,4,6-tris(dimethyl aminomethyl) phenol, morpholine, Amine compounds, such as N-methylmorpholine, 2-ethyl-4-methylimidazole, 1, and 8-diazabicyclo (5.4.0) undecene 7 (DBU), or salt [with carboxylic acid of these amine compounds, etc.]; — low-molecular-weight-polyamide resin; obtained from superfluorous polyamine and polybasic acid — resultant; gamma-aminopropyl trimethoxysilane of superfluorous polyamine and an epoxy compound. Publicly known silanol condensation catalysts, such as silanol condensation catalysts, such as silane coupling agent, which has amino groups, such as N-(beta-aminoethyl) aminopropyl methyl dimethoxysilane, an acid catalyst of further others, and a basic catalyst, etc. are mentioned. These catalysts may be used alone and may be used together two or more sorts.

[0029]As for the amount of these curing catalysts used, about 0.1–20 copies are preferred to 100 copies of reactive silicon group content oxypropylene polymers, and its about 1–10 copies are still more preferred. Since a cure rate will become slow and a hardening reaction will become fully difficult to advance to a reactive silicon group content oxypropylene polymer if there is too little amount of curing catalyst used, it is not desirable. On the other hand, if there is too much amount of curing catalyst used to a reactive silicon group content oxypropylene polymer, since a good hardened material will become generation of heat and foaming local at the time of hardening arise, and is hard

to be obtained, it is not desirable.

[0030]The reactive silicon group content oxypropylene polymer can denaturalize by mixing various bulking agents. As a bulking agent, fumes silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, and reinforcement nature bulking agent, calcium carbonate like carbon black, Magnesium carbonate, diatomite, calcination clay, clay, talc, titanium oxide, **** bulking agents, such as bentonite, organic bentonite, ferric oxide, a zinc oxide, an active white, hydrogenation castor oil, and a milt balloon; asbestos, glass fiber, and a fibrous filler like a filament are illustrated.

[0031]To obtain a strong high hardening constituent with these bulking agents. Mainly Fumes silica, sedimentation nature silica, a silicic acid anhydride, hydrous silicic acids, carbon black, A desirable result will be obtained if a bulking agent chosen from surface treatment detailed calcium carbonate, calcination clay, clay, an active white, etc. is used in 1–100 copies to 100 copies of reactive silicon group content oxypropylene polymers. When elongation wants to obtain a hardening constituent which is size with low strength, A desirable result will be obtained if a bulking agent mainly chosen from titanium oxide, calcium carbonate, magnesium carbonate, talc, ferric oxide, a zinc oxide, a milt balloon, etc. is used in 5–200 copies to 100 copies of reactive silicon group content oxypropylene polymers. Of course, these bulking agents may be used only by one kind, and may carry out two or more kind mixing use.

[0032]In a hardenability constituent of this invention, since elongation of a hardened material can be enlarged or a lot of bulking agents can be mixed if a plasticizer is used, using it together with a bulking agent, it is more effective. As this plasticizer, dioctyl phthalate, dibutyl phthalate, **** phthalic ester, such as butylbenzyl phthalate; Dioctyl adipate, **** aliphatic dibasic acid ester, such as succinic acid isodecyl and dibutyl sebacate; Diethylene glycol dibenzoate, **** glycol ester, such as pentaerythritol ester; Butyl oleate, **** aliphatic series ester species, such as methyl acetyl ricinolate; Tricresyl phosphate, **** phosphoric ester, such as triethyl phosphate and phosphoric acid octyldiphenyl; Epoxidized soybean oil, **** epoxy plasticizer [such as epoxy stearic acid benzyl]; — polyester plasticizer [such as polyester of dibasic acid and dihydric alcohol]; — polyether [such as a polypropylene glycol and its derivative]; — Poly alpha-methylstyrene. Polystyrene, such as polystyrene; plasticizers, such as polybutadiene, Butadiene Acrylonitrile, polychloroprene, polyisoprene, polybutene, and chlorinated paraffins, can use it independently arbitrarily in a form of two or more kinds of mixtures. A desirable result will be obtained if the amount of plasticizers is used in 0–100 copies to 100 copies of reactive silicon group content oxypropylene polymers.

[0033]An ingredient which limitation in particular does not have in preparation of a hardenability constituent of this invention, for example, was described above is blended, it kneads under ordinary temperature or heating using a mixer, a roll, a kneader, etc., or an ingredient is dissolved using a little suitable solvents, a usual method of mixing is adopted, and it gets. 1 liquid type and a two-component compound can also be built and used by combining these ingredients suitably.

[0034]If a hardenability constituent of this invention is exposed into the atmosphere, by operation of moisture, it will form network structure in three dimensions, and will harden it to a solid which has rubber-like elasticity.

[0035]It faces using a hardenability constituent of this invention, and it is still more possible to add suitably various additive agents, such as an adhesive improving agent, a physical-properties regulator, a preservation stability improving agent, lubricant, paints, and a foaming agent, if needed.

[0036]Especially a hardenability constituent of this invention is useful as elastic sealant, and can be used as seal agents, such as a building, a marine vessel, a car, and a road. Since, or help of a primer is borrowed and it may stick to substrates of a **** large area, such as glass, porcelain, wood, metal, and a resin-molding thing, it is usable also as various seal constituents and adhesion constituents of a type, it is useful also as a food packaging material, a casting rubber material, a charge of templating material, and a paint.

[0037]

[Effect of the Invention]The constituent of this invention becomes the thing excellent in the weatherability and heat resistance of a hardened material as compared with the constituent which used the large polymer of molecular weight distribution as a (A) ingredient.

[0038]Although the reactive silicon group content oxypropylene polymer used as a (A) ingredient in the hardenability constituent of this invention has a large number average molecular weight, its

molecular weight distribution is narrow. Therefore, compared with the constituent which contains the conventional reactive silicon group content oxypropylene polymer with large molecular weight distribution with the same molecular weight before hardening, viscosity is low and the constituent of this invention is easy handling.

[0039] Thus, since the viscosity before hardening is low, the room-temperature-curing nature constituent which workability could blend a lot of bulking agents, and it is not only good, but was excellent can be obtained.

[0040] Chemical resistance, such as acid resistance, is improved substantially unexpectedly, and the water resisting property is also excellent.

[0041]

[Example] In order to clarify this invention further, working example is hung up over below.

[0042] 401 g (0.081 Eq) of polyoxypropylene triol (Mw/Mn=1.38, viscosity of 89 poise) of the molecular weight 15,000 was taught to synthetic 1.5 l. of example resisting pressure glass reaction vessels, and it carried out under a nitrogen atmosphere.

[0043] Decompression devolatilization was carried out, after dropping the 28% methanol solution 19.1 g (0.099 Eq) of sodium methoxide and making it react from a tap funnel at 137 °C for 5 hours. It returned under a nitrogen atmosphere, and the allyl chloride 9.0 g (0.118 Eq) was further arylated using the 28% methanol solution 5.6 g (0.029 Eq) and the allyl chloride 2.7 g (0.035 Eq) of sodium methoxide, after making it react for 1.5 hours, dropping and.

[0044] If decompression removal of the hexane is carried out after melting this reactant in hexane and carrying out adsorption treatment with aluminum silicate — the yellow of 311 g — transparent polymer was obtained (viscosity of 68 poise).

[0045] 270 g (0.065 Eq) of this polymer was taught to resisting pressure glass reaction vessels, and it carried out under a nitrogen atmosphere. It agitated for 30 minutes after adding 0.075 ml of catalyst solutions (solution which melted H_2PtCl_6 and $6H_2O$ 25g in the isopropyl alcohol 500g) of chloroplatinic acid, if it devolatilizes after adding 6.24 g (0.059 Eq) of dimethoxymethylsilane from a tap funnel and making it react at 90 °C for 4 hours — the yellow of 260 g — transparent polymer was obtained.

[0046] 220 g (0.0447 Eq) of polyoxypropylene triol (Mw/Mn=1.38, viscosity of 89 poise) and 0.02 g of diallaic acid dibutyltin of the number average molecular weight 15,000 are taught to a flask with synthetic example 2 agitator, 8.45 g (0.0447 Eq) of gamma-isocyanate propylmethyl dimethoxysilane was dropped at the room temperature under a nitrogen atmosphere. It was made to react at 75 °C after the end of dropping for 1.5 hours. The reaction was terminated, after measuring an IR spectrum and checking disappearance of the NCO absorption near 2280 cm^{-1} , and generation of the C=O absorption near 1730 cm^{-1} . 213 g of water-white polymer was obtained.

[0047] Example of comparison composition 1 number average molecular weight taught the resisting pressure glass reaction vessels in which the nitrogen purge of the polyoxypropylene glycol 420g and the number average molecular weight of 3,000 was carried out in 80 g of polyoxypropylene triol of 3,000. After adding 40 g of sodium hydroxide and making it react at 60 °C for 13 hours, 19 g of bromo methyl chlorides were made to react at 60 °C for 10 hours, (Mw/Mn of the obtained polymer was 2.1 and viscosity was 385 poise).

Then, the allyl chloride 15g was added and the reaction was performed for 36 hours. After ending reaction, decompression was used and the volatile substance was removed.

[0048] Contents were taken out to the beaker and it melted in hexane. After carrying out adsorption treatment with aluminum silicate, decompression removal of the hexane was carried out.

[0049] After teaching 500 g of this polymer to the reaction vessel by which the nitrogen purge was carried out and adding 0.03 g of catalyst solutions (solution which melted H_2PtCl_6 and $6H_2O$ 25g in the isopropyl alcohol 500g) of chloroplatinic acid, 12 g of dimethoxymethylsilane was added and it was made to react at 80 °C for 4 hours. If decompression is used and a volatile substance is removed after ending reaction — light yellow — 550g of transparent polymer was obtained.

[0050] The viscosity of the polymer obtained in the synthetic examples 1 and 2 and the example 1 of comparison composition was measured at 23 °C using the Brookfield viscometer (BM type rotor No.4, 12 rpm). GPC analyzed the number average molecular weight (Mn) and molecular weight distribution (Mw/Mn) of each polymer. GPC used the tetrahydrofuran for the column filled up with polystyrene gel

(made by TOSOH CORP.) as a distillate solvent, and analyzed it with the oven temperature of 40 °C. The result is shown in Table 1.

[0051]

[Table 1]

重合体	粘度 (ポイズ)	数平均分子量 (Mn)	分子重量分布 (Mw/Mn)
合成例 1	88	1.8×10^4	1.5
合成例 2	150	1.7×10^4	1.4
比較合成例 1	380	1.8×10^4	2.3

[0052] As opposed to 100 copies of polymer obtained in working example 1 and the example 1 of comparative example 1 composition, or the example 1 of comparison composition, NOCRAC SP made from Ouchi Shinko Chemical Industry as a hindered phenolic antioxidant One copy, SANORU LS770 by Sankyo Co., Ltd. was added as one copy and a hindered amine system antioxidant, three copies of octylic acid tin and 0.5 copy of lauryl amine were added as one copy and a curing catalyst, and TINUVIN327 made from CIBA-GEIGY was kneaded uniformly. Among the obtained constituents, the constituent (thing using polymer of the synthetic example 1) of working example 1 had low viscosity compared with the constituent (thing using polymer of the example 1 of comparison composition) of the comparative example 1, and was easy handling.

[0053] After creating a 3-mm-thick sheet using these constituents, it was recuperated for three days at 50 more °C for two days at 23 °C. When the sunshine WOM estimated this hardening constituent, by it, the surface dissolved the constituent of working example 1 for a while 720 hours afterward. On the other hand, the surface dissolved the constituent of the comparative example 1 for a while 480 hours afterward.

[0054] To 100 copies of polymer obtained in the example 1 of reference example, composition, or the example 1 of comparison composition, three copies of octylic acid tin and 0.5 copy of lauryl amine were added, and it kneaded uniformly.

[0055] After creating a 3-mm-thick sheet using these constituents, it was recuperated for three days at 50 more °C for two days at 23 °C. When the sunshine WOM estimated this hardening constituent, by it, the surface dissolved all for a while 72 hours afterward.

[0056] It replaced with the polymer obtained in the example 1 of working example 2 composition, and the hardenability constituent was obtained like working example 1 using the polymer obtained in the synthetic example 2. The hardened material of this constituent had the same outstanding weatherability as the thing of working example 1.

[Translation done.]